

# Symmetry classification of bond order parameters in cuprates

Roland Zeyher

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

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We study bond-order parameters for generalized  $t$ - $J$  models on a square lattice. Using the plane-wave limit the considered order parameters form basis functions for irreducible representations of the symmetry transformations of the point group and of time reversal. We show that for instability wave vectors along the diagonals all possible basis functions are either fine-tuned (i.e., obey restrictions beyond the requirements of symmetry) or break time reversal symmetry and thus describe flux states. For instability wave vectors along the crystalline axes, corresponding to the observed case in underdoped cuprates, there are only three representations with  $A_1$ ,  $B_1$ , and  $E$  symmetry which do not break time reversal symmetry in the general case. We suggest that one of them has recently been observed in resonant elastic X-ray scattering.

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## I. INTRODUCTION

There is growing evidence from nuclear magnetic resonance<sup>1</sup>, resonant X-ray scattering and diffraction<sup>2-8</sup> and scanning tunneling microscopy<sup>8-10</sup> that charge ordered states play an important role in underdoped cuprates. In particular, a charge-modulated state with 4 incommensurate wave vectors along the crystalline axes was detected in YBCO and in Bi-based single and double layer compounds. Resonant elastic X-ray scattering showed that the charge order emerges just below the opening of the pseudogap<sup>11</sup> in underdoped Bi2201<sup>9</sup>. The formation of the pseudogap, Fermi pockets, the appearance of quantum oscillations<sup>12</sup> and of charge order may thus be intimately related in these systems. It is the aim of this paper to characterize charge ordered states in interacting systems independently of the strength of the interaction. Furthermore, the implications of point group and time reversal symmetries as well as the Hermiticity of the Hamiltonian for the order parameter (OP) will be taken into account in greater detail than in previous treatments.

The microscopic form of the charge OP in cuprates is not clear at present. To illustrate this let us consider the following model Hamiltonian for electrons on a square lattice which generally is believed to be relevant for cuprates<sup>13</sup>,

$$H = - \sum_{i,j} t_{ij} c_{i,\alpha}^\dagger c_{j,\alpha} + \frac{J}{4} \sum_{\langle i,j \rangle} c_{i,\alpha}^\dagger c_{i,\beta} c_{j,\beta}^\dagger c_{j,\alpha} + \frac{V}{2} \sum_{\langle i,j \rangle} c_{i,\alpha}^\dagger c_{i,\alpha} c_{j,\beta}^\dagger c_{j,\beta}. \quad (1)$$

$t_{ij}$  denotes the hopping amplitude of electrons between the lattice site  $i$  and  $j$ .  $c_{i,\alpha}^\dagger$  and  $c_{i,\alpha}$  are fermionic creation and annihilation operators,  $\alpha$  spin indices and repeated spin indices are always summed over. The second and third terms in Eq. (1) describe antiferromagnetic and Coulomb interactions between electrons on neighboring sites  $i$  and  $j$  with coupling constants  $J$  and  $V$ , respec-

tively. If double occupancies of sites are excluded Eq. (1) represents the well-known  $t$ - $J$  model for  $V = -J/2$ .

The interaction terms in Eq. (1) give rise to two kinds of charge OPs. A Hartree-like contraction of the third term yields an OP proportional to  $\langle c_{i\alpha}^\dagger c_{i\alpha} \rangle$  describing the charge on the site  $i$ . It may vary from site to site and represents a conventional charge density wave (CDW) state. The exchange contractions of the second and third term in Eq. (1) yield an OP proportional to  $\langle c_{i\alpha}^\dagger c_{j\alpha} \rangle$ , where  $i$  and  $j$  are nearest neighbor sites. This state may be called a nonlocal CDW or a bond-order wave (BOW) state<sup>14</sup> where the CDW acquires an internal degree of freedom because the electron and hole occupy different sites. It has been shown that in the large  $N$  limit of the  $t$ - $J$  model (which corresponds to enforcing the constraint of no double occupancies of sites only globally) the phase diagram consists in the underdoped regime of incommensurate BOW states (at zero doping of the staggered flux phase<sup>15</sup> as a special case)<sup>16,17</sup>. At the same time the conventional CDW OP is zero showing that both kinds of charge order are independent from each other. More recently the BOW state has been studied theoretically in more detail<sup>18-22</sup>. Also models with more than one band<sup>23</sup> or more complex OPs<sup>22,24</sup> have been considered. Recently a microscopic form for the OP in underdoped YBCO and Bi2201 was proposed<sup>8</sup> based on experimental data from resonant X-ray scattering.

Throughout the paper we will assume that the temperature is below the transition temperature to the BOW state. The OPs are then in general nonzero and their symmetry properties can be studied. We will classify possible OPs for BOW states by exploiting point-group and time reversal symmetries as well as the Hermiticity of  $H$ . In the appendix it is shown that possible OPs for the ground state are basis functions for representations of  $C_{4v}$ . If the ground state is non-degenerate in the sense that it does not contain two linearly independent OPs the representation is irreducible. If the ground state is degenerate and satisfies a two dimensional representation this representation may be irreducible or reducible. In the latter case it is composed of two OPs with differ-

ent symmetries and the degeneracy is not a consequence of symmetry but of coupling constants. In the following we will confine our discussion to OPs which form irreducible representations of  $C_{4v}$  and exclude accidental degeneracies, additional instabilities or induced higher harmonics<sup>25</sup>. Explicit expressions for the OPs will be given for BOW states with four wave vectors of the form  $(\pm q, \pm q)$  and the form  $(\pm q, 0)$  and  $(0, \pm q)$ .

## II. DEFINITION AND TRANSFORMATION PROPERTIES OF THE ORDER PARAMETER

From Eq. (1) follows that the BOW OP has the form of a coupling constant times the matrix element  $\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle$ , where  $i$  and  $j$  are nearest neighbors. To simplify the nomenclature we will call the modulated part of  $\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle$  OP in the following. After a Fourier transform we obtain,

$$\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \sum_{\mathbf{k}, \mathbf{q}} \langle c_{\mathbf{k}+\mathbf{q},\alpha}^\dagger c_{\mathbf{k},\alpha} \rangle e^{-i\mathbf{q}\mathbf{r}_i} e^{-i(\mathbf{r}_i-\mathbf{r}_j)\cdot\mathbf{k}}. \quad (2)$$

$\mathbf{r}_i$  is the vector from the origin to the lattice site  $i$ . The sum over  $\mathbf{q}$  in Eq. (2) includes in the plane-wave limit only the wave vectors corresponding to a charge instability of the normal state. They form a star of wave vectors  $\{\mathbf{q}_l\}$ . Writing  $\mathbf{r}_j = \mathbf{r}_i + \mathbf{e}_j$ , keeping  $\mathbf{e}_j$  fixed and performing a Fourier transformation with respect to  $\mathbf{r}_i$  we get

$$\sum_{\mathbf{r}_i} \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle e^{i\mathbf{q}_l \mathbf{r}_i} = \sum_{\mathbf{k}} \langle c_{\mathbf{k}+\mathbf{q}_l,\alpha}^\dagger c_{\mathbf{k},\alpha} \rangle e^{i\mathbf{e}_j \cdot \mathbf{k}} = F(\mathbf{q}_l, \mathbf{e}_j). \quad (3)$$

The functions  $F(\mathbf{q}_l, \mathbf{e}_j)$  are defined by Eq. (3) and represent our set of order parameters. If Umklapp terms are included the sum over  $\mathbf{q}_l$  may not only include primary instability vectors of the normal state but higher harmonics with wave vectors  $\sum_l n_l \mathbf{q}_l$  where  $n_l$  is an integer. They form new stars and cause deviations from the plane-wave limit of the OP. Because these higher harmonics are important only near the transition to the commensurate phase we will neglect them in the following and restrict ourselves to the plane-wave limit.

The symmetry group of the square lattice is  $C_{4v}$ . Using the notation of Ref.<sup>25</sup> let us denote one of the 8 symmetry transformations by  $R$ . Its action on the order parameter in Eq. (3) can be written as

$$\langle c_{\mathbf{R}^{-1}\mathbf{r}_i,\alpha}^\dagger c_{\mathbf{R}^{-1}\mathbf{r}_j,\alpha} \rangle = \sum_{\mathbf{k}, l} \langle c_{\mathbf{k}+\mathbf{R}^{-1}\mathbf{q}_l,\alpha}^\dagger c_{\mathbf{k},\alpha} \rangle e^{-i\mathbf{q}_l \cdot \mathbf{r}_i} e^{i\mathbf{k} \cdot \mathbf{R}^{-1}\mathbf{e}_j}, \quad (4)$$

where  $\mathbf{R}$  is the 2x2 matrix representing  $R$  in the two-dimensional direct space. After a Fourier transformation with respect to  $\mathbf{r}_i$  we find that  $F(\mathbf{q}_l, \mathbf{e}_j)$  transforms under  $R$  into  $F(\mathbf{R}^{-1}\mathbf{q}_l, \mathbf{R}^{-1}\mathbf{e}_j)$ , where  $\mathbf{R}^{-1}\mathbf{q}_l$  and  $\mathbf{R}^{-1}\mathbf{e}_j$  belong to the star of wave vectors and to nearest neighbor bonds, respectively. This means that the set of functions  $\{F(\mathbf{q}_l, \mathbf{e}_j)\}$  forms basis functions for a (reducible) representation of  $C_{4v}$ . Decomposing this reducible representation into irreducible parts the basis function of one of the

irreducible representations describes the OP of the state corresponding to the global minimum of the free energy. One important feature is that in general both  $\mathbf{q}_l$  and  $\mathbf{e}_j$  are transformed under  $R$  at the same time and not independently from each other. This is a crucial point in our approach.

Further general properties of the functions  $\{F(\mathbf{q}_l, \mathbf{e}_j)\}$  are related to time reversal and the Hermiticity of the Hamiltonian. The operator  $T$  for time reversal is defined in real space by

$$T \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle^*, \quad (5)$$

where the star means conjugate complex. Taking Fourier transforms on both sides we get,

$$\sum_{\mathbf{r}_i} e^{-i\mathbf{q}_l \mathbf{r}_i} T \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \sum_{\mathbf{r}_i} e^{-i\mathbf{q}_l \mathbf{r}_i} \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle^*, \quad (6)$$

or

$$T \sum_{\mathbf{r}_i} e^{i\mathbf{q}_l \mathbf{r}_i} \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle (\sum_{\mathbf{r}_i} e^{i\mathbf{q}_l \mathbf{r}_i} \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle)^*, \quad (7)$$

or,

$$TF(\mathbf{q}_l, \mathbf{e}_j) = (F(\mathbf{q}_l, \mathbf{e}_j))^* = F^*(\mathbf{q}_l, \mathbf{e}_j). \quad (8)$$

In Eqs. (6) and (7)  $c_{j,\alpha}$  stands for  $c_{\mathbf{r}_i+\mathbf{e}_j,\alpha}$  and  $\mathbf{e}_j$  is fixed in the sums over  $\mathbf{r}_i$ .  $F^*(\mathbf{q}_l, \mathbf{e}_j)$  is defined by the last equation. Some authors interpret the right-hand side of Eq. (6) as a Fourier transform of  $\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle$  which may be written as  $(F^*)(\mathbf{q}_l, \mathbf{e}_j)$ . The connection to our definition Eq. (8) is  $F^*(\mathbf{q}_l, \mathbf{e}_j) = (F^*)(-\mathbf{q}_l, \mathbf{e}_j)$ . In order to avoid confusion we will always use our definition in Eq. (8). Using the Hermiticity of the Hamiltonian the second half of Eq. (3) yields

$$F^*(\mathbf{q}_l, \mathbf{e}_j) = e^{i\mathbf{e}_j \cdot \mathbf{q}_l} F(-\mathbf{q}_l, -\mathbf{e}_j) = e^{i\mathbf{e}_j \cdot \mathbf{q}_l} C_2 F(\mathbf{q}_l, \mathbf{e}_j). \quad (9)$$

$C_2$  denotes the rotation by  $\pi$ .

Eq. (8) implies  $T^2 = 1$ , which corresponds to the case of integral spin.<sup>25</sup> Applying the Frobenius-Schur test<sup>25</sup> to the point group  $C_{4v}$  shows that including  $T$  in the set of symmetry transformations cannot produce additional degeneracies of irreducible representations. Thus it is convenient to construct first basis functions for irreducible representations of the point group and then to check their behavior under time reversal.

## III. WAVE VECTORS OF THE BOW ALONG THE DIAGONALS

In the following we will first consider the case with 4 wave vectors along the diagonals, i.e.,  $\mathbf{q}_1 = (q, q)$ ,  $\mathbf{q}_2 = (-q, q)$ ,  $\mathbf{q}_3 = (-q, -q)$ ,  $\mathbf{q}_4 = (q, -q)$ , where  $q$  lies between 0 and  $\pi$ . The four bond directions are denoted by  $\mathbf{e}_1 = (1, 0)$ ,  $\mathbf{e}_2 = (0, 1)$ ,  $\mathbf{e}_3 = (-1, 0)$ ,  $\mathbf{e}_4 = (0, -1)$ . It is easy to see that the following 8 functions  $F_l =$

TABLE I: Coefficients  $c_{\gamma l}$ 

$\gamma$	$F_1$	$F_2$	$F_3$	$F_4$	$F_5$	$F_6$	$F_7$	$F_8$
$A_1$	1	1	1	1	1	1	1	1
$A_2$	1	1	1	1	-1	-1	-1	-1
$B_1$	1	-1	1	-1	1	1	-1	-1
$B_2$	1	-1	1	-1	-1	-1	1	1
$E^{(1)}(1)$	1	-1	-1	1	1	-1	-1	1
$E^{(1)}(2)$	1	1	-1	-1	1	-1	1	-1
$E^{(2)}(1)$	1	-1	-1	1	-1	1	1	-1
$E^{(2)}(2)$	1	1	-1	-1	-1	1	-1	1

$F(\mathbf{q}_l, \mathbf{e}_l), l = 1, \dots, 4$ ,  $F_5 = F(\mathbf{q}_4, \mathbf{e}_1)$ ,  $F_6 = F(\mathbf{q}_2, \mathbf{e}_3)$ ,  $F_7 = F(\mathbf{q}_1, \mathbf{e}_2)$ , and  $F_8 = F(\mathbf{q}_3, \mathbf{e}_4)$  yield a reducible representation of  $C_{4v}$ . Let us denote the linear combinations of the  $F_l$  which form basis functions for the corresponding irreducible representations  $\gamma$  by

$$F(\gamma) = \sum_{l=1}^8 c_{\gamma l} F_l. \quad (10)$$

For the one-dimensional representations  $\gamma = A_1, A_2, B_1, B_2$  one can easily determine the coefficients  $c_{\gamma l}$  from the character table of  $C_{4v}$ . One finds that each of these representations occurs exactly one time, the corresponding  $c_{\gamma l}$  are given in the first 4 lines of Table 1. Using again the character table one finds that the remaining 4 functions form 2 two-dimensional representations  $E^{(1)}$  and  $E^{(2)}$ . The corresponding  $c_{\gamma l}$  are given in the lines 5-8 in Table 1.

Going back to Eq. (9) we note that for the functions  $F_l, l = 1, \dots, 8$  the phase factor  $\mathbf{e}_j \cdot \mathbf{q}_l$  is always equal to  $q$ . Multiplying Eq. (9) by  $c_{\gamma l}$  and summing over  $l$  we obtain

$$F^*(\gamma) = e^{iq} C_2 F(\gamma). \quad (11)$$

From the character table of  $D_4$  follows that  $C_2 F(\gamma)$  is equal to  $F(\gamma)$  for  $\gamma = A_1, A_2, B_1, B_2$  and equal to  $-F(\gamma)$  for  $\gamma = E$ . The solution of Eq. (11) is

$$F(\gamma) = \left(1 + i \frac{\cos q \mp 1}{\sin q}\right) \text{Re} F(\gamma). \quad (12)$$

where the upper sign refers to  $\gamma = A_1, A_2, B_1, B_2$  and the lower sign to  $\gamma = E$ , respectively. The real part of  $F(\gamma)$ ,  $\text{Re} F(\gamma)$ , may assume any real number.

For  $q = 0$  or  $\pi$  the four functions  $F_1, \dots, F_4$  form a basis for a reducible representation of  $C_{4v}$  which decomposes into  $A_1, B_1$  and  $E$  representations with the basis functions  $F_1 + F_2 + F_3 + F_4$ ,  $F_1 - F_2 + F_3 - F_4$  and  $F_1 - F_3, F_2 - F_4$ , respectively. Since Eq. (11) still holds the first two basis functions are real (imaginary) and the third and fourth ones imaginary (real) for  $q = 0$  ( $q = \pi$ ). Included as a special case is the staggered flux phase with wave vector  $(\pi, \pi)$ . It has  $B_1$  symmetry and a purely imaginary OP in agreement with previous conclusions.<sup>15-19</sup>

Let us denote the second set of 8 functions by  $\tilde{F}_l, l = 1, \dots, 8$ . Each  $\tilde{F}_l$  is obtained from  $F_l$  by exchanging  $\mathbf{e}_1$

with  $\mathbf{e}_3$  and  $\mathbf{e}_2$  with  $\mathbf{e}_4$ . The linear space spanned by the functions  $\tilde{F}_l = 1, \dots, 8$  yields a reducible representation of  $C_{4v}$ . Decomposing it into its irreducible parts gives one time the representations  $A_1, A_2, B_1, B_2$  and two times the representation  $E$ , exactly as for the first 8 functions  $F_l$ . The analogue of Eq. (10) reads

$$\tilde{F}(\gamma) = \sum_{l=1}^8 c_{\gamma l} \tilde{F}_l, \quad (13)$$

where the coefficients  $c_{\gamma l}$  are the same as in Table 1. The phase factor  $\mathbf{e}_j \cdot \mathbf{q}_l$  is for all 8 functions equal to  $-q$  so that Eq. (11) reads

$$\tilde{F}^*(\gamma) = e^{-iq} C_2 \tilde{F}(\gamma), \quad (14)$$

with the solution

$$\tilde{F}(\gamma) = \left(1 - i \frac{\cos q \mp 1}{\sin q}\right) \text{Re} \tilde{F}(\gamma). \quad (15)$$

$\text{Re} \tilde{F}(\gamma)$  is unrelated to  $\text{Re} F(\gamma)$  and may assume any real value. This expresses the fact that  $F$  and  $\tilde{F}$  describe possible OPs for all values of  $\text{Re} F(\gamma)$  and  $\text{Re} \tilde{F}(\gamma)$ .

Complex functions for  $F(\gamma)$  and  $\tilde{F}(\gamma)$  do not necessarily imply that the corresponding states break  $T$  symmetry. This is true in particular in our case because a possible imaginary part to  $F$  can come from the matrix element but also from combinations of the exponential functions in Eq. (3). A general criterion for an unbroken  $T$  symmetry follows from Eq. (5), namely,

$$T \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle. \quad (16)$$

Applying Fourier transformations on both sides of Eq. (16) similar as in Eqs. (6) and (7) yields

$$TF(\mathbf{q}_l, \mathbf{e}_j) = F(-\mathbf{q}_l, \mathbf{e}_j). \quad (17)$$

Comparing with Eq. (8) yields the following criterion which must be fulfilled if  $T$  symmetry is unbroken,

$$F(-\mathbf{q}_l, \mathbf{e}_j) = F^*(\mathbf{q}_l, \mathbf{e}_j). \quad (18)$$

For the special case  $\mathbf{q}_l = 0$  the above criterion is fulfilled for the real OP of  $A_1$  and  $B_1$  symmetry, found above, but not for the imaginary OP of the  $E$  symmetry. Thus  $T$  symmetry is unbroken for the  $A_1$  and  $B_1$  and broken for the  $E$  symmetry. For  $\mathbf{q}_l = (\pi, \pi)$  one finds that the purely imaginary OPs of the  $A_1, B_1$  symmetries break and the real OP of the  $E$  symmetry preserves  $T$  symmetry.

Considering a general  $\mathbf{q}_l$  along the diagonals, a ground state with symmetry  $\gamma$  is in general given by a linear combination of all basis functions belonging to the same representation. It has thus in our case the form  $\alpha F(\gamma) + \beta \tilde{F}(\gamma)$  with coefficients  $\alpha$  and  $\beta$  which have to be real to be compatible with Eq. (12) and (15). Applying  $T$  to this state we obtain,

$$T(\alpha F(\gamma) + \beta \tilde{F}(\gamma)) = \alpha TF(\gamma) + \beta T\tilde{F}(\gamma). \quad (19)$$

Inserting the functions  $F_l$  into Eq. (8), multiplying by  $c_{\gamma l}$ , summing over  $l$  and using Eq. (11) yields

$$TF(\gamma) = F^*(\gamma) = e^{iq}C_2F(\gamma). \quad (20)$$

Replacing  $F_l$  by  $\tilde{F}_l$  and using Eq. (14) instead of Eq. (11) gives

$$T\tilde{F}(\gamma) = \tilde{F}^*(\gamma) = e^{-iq}C_2\tilde{F}(\gamma). \quad (21)$$

Eqs. (20) and (21) always hold. If, in addition,  $T$  symmetry is preserved we obtain from Eq. (17),

$$TF(\gamma) = C_2\tilde{F}(\gamma), \quad (22)$$

and

$$T\tilde{F}(\gamma) = C_2F(\gamma). \quad (23)$$

Inserting Eqs. (20) - (23) into Eq. (19) yields

$$\tilde{F}(\gamma) = e^{iq}F(\gamma). \quad (24)$$

Noting that Eqs. (12) and (15) can also be written as

$$F(\gamma) = e^{-iq/2} / \cos(q/2) \cdot \text{Re}F(\gamma) \quad (25)$$

and

$$\tilde{F}(\gamma) = e^{iq/2} / \cos(q/2) \cdot \text{Re}\tilde{F}(\gamma), \quad (26)$$

Eq. (24) is equivalent to

$$\text{Re}F(\gamma) = \text{Re}\tilde{F}(\gamma), \quad (27)$$

or, using Eqs. (10) and (13),

$$\sum_l c_{\gamma l} \text{Re}(F_l) = \sum_l c_{\gamma l} \text{Re}(\tilde{F}_l). \quad (28)$$

The functions  $F_l$  and  $\tilde{F}_l$  are different from each other and the functions of each of the two sets transform within each set under the elements of the point group and under time reversal. Thus symmetry does not enforce any relation between  $\text{Re}F(\gamma)$  and  $\text{Re}\tilde{F}(\gamma)$  and Eq. (27) will not necessarily be fulfilled for a general Hamiltonian. However, this does not exclude OPs exhibiting  $T$  symmetry. The left and right-hand sides of Eqs. (27) may assume independently any real value. The case where both values are equal is not excluded and represents an OP with  $T$  symmetry. Such a fine-tuned state may, however, be vulnerable to perturbations, for instance, to a change in the coupling constants, the temperature etc. Since no general argument seems to exist which protects Eq. (27) against such perturbations it is reasonable to conclude that in the general case the basis functions in Eq. (10) and Eq. (13) break  $T$  symmetry for  $\mathbf{q} \neq 0$  and  $\neq (\pi, \pi)$ . Whether for a specific Hamiltonian the ground state breaks or preserves  $T$  symmetry can only be determined by an explicit calculation of the OPs and the free energy. In the next section we will encounter a totally different case where the breaking and preserving of  $T$  symmetry is enforced by symmetry independently of the values of microscopic coupling constants or specific Hamiltonians.

#### IV. WAVE VECTORS OF THE BOW ALONG THE AXES

Next we consider the case of four wave vectors along the crystalline axes, i.e.,  $\mathbf{q}_1 = (q, 0)$ ,  $\mathbf{q}_2 = (0, q)$ ,  $\mathbf{q}_3 = (-q, 0)$ , and  $\mathbf{q}_4 = (0, -q)$  where  $q$  lies between 0 and  $\pi$ . Using the previous notation the functions  $F_l$ ,  $l = 1, \dots, 4$  yield irreducible representations of  $A_1, B_1$  and  $E$  symmetries with the basis functions  $F_1 + F_2 + F_3 + F_4$ ,  $F_1 - F_2 + F_3 - F_4$ ,  $F_1 - F_3$ , and  $F_2 - F_4$ , respectively. These basis functions can be written in the form of Eq. (10) where the sum runs from 1 to 4 and the corresponding  $\gamma$  in Table 1 is chosen. The exponential factor in Eq. (9) is in each case  $e^{iq}$ . Thus Eq. (11) holds for these three representations and their basis functions are complex for  $q \neq 0$ . All the above results also apply to the set  $\tilde{F}_l$ ,  $l = 1, \dots, 4$ , if the exponential factor is replaced by  $e^{-iq}$ . Thus each of the manifolds  $F$  and  $\tilde{F}$  lead to one  $A_1, B_1$  and one  $E$  representation and their basis functions are complex for  $q \neq 0$ . The arguments concerning  $T$  breaking in Eqs. (19) - (28) can be transferred to the present star of wave vectors with the result that all states which are not fine-tuned in the sense discussed above break  $T$  symmetry.

The remaining 8 functions are conveniently split into the combinations  $F_l^+ = F(\mathbf{q}_l, \mathbf{e}_{l+1}) + F(\mathbf{q}_l, \mathbf{e}_{l+3})$  and  $F_l^- = F(\mathbf{q}_l, \mathbf{e}_{l+1}) - F(\mathbf{q}_l, \mathbf{e}_{l+3})$ , where  $\mathbf{e}_5, \mathbf{e}_6$  etc. mean  $\mathbf{e}_1, \mathbf{e}_2$  etc. and  $l$  runs from 1 to 4. The functions  $F_l^+, l = 1, \dots, 4$  lead to  $A_1, B_1$  and  $E$ , the functions  $F_l^-, l = 1, \dots, 4$  lead to  $A_2, B_2$  and  $E$  representations. The corresponding basis functions are the same as for the above  $F_l$  manifold, once  $F_l$  is replaced by  $F_l^+$  or  $F_l^-$ , respectively. For all 8 functions the wave and the bond vectors are perpendicular to each other implying that  $q$  is identical to zero and that no complex exponential appears in Eq. (9). As a result we get  $F^{+*}(\gamma) = C_2F^+(\gamma)$  so that  $F^+(\gamma)$  is real for  $\gamma = A_1, B_1$  and imaginary for  $\gamma = E$ . Using these properties we have from Eq. (8)

$$TF^+(\gamma) = C_2F^+(\gamma), \quad (29)$$

for  $l = 1, \dots, 4$ . Eq. (29) is a direct consequence of the definition of the operator  $T$  and holds in any case. If in addition  $T$  symmetry applies Eq. (17) also holds. Forming appropriate basis functions Eq. (17) is identical with Eq. (29) after taking into account that  $F^+(A_1)$  and  $F^+(B_1)$  are real and  $F^+(E)$  imaginary. This means that the condition for  $T$  symmetry is automatically fulfilled in this case for all three representations. Remarkable is that no condition of the kind of Eq. (24) or Eq. (27) appears which cannot be fulfilled in the general case. The  $T$  symmetry arises here without fine-tuning and is enforced by symmetry.

Finally, let us consider the basis functions  $F^-(\gamma)$  of the irreducible representations  $\gamma = A_2, B_2, E$ . Eq. (9) yields  $F^{-*}(\gamma) = C_2F^-(\gamma)$  implying that  $F^-(A_2)$  and  $F^-(B_2)$  are real and  $F^-(E)$  are imaginary. Forming basis func-

tions in Eqs. (8) and (9) gives

$$TF^-(\gamma) = C_2 F^-(\gamma). \quad (30)$$

If in addition  $T$  symmetry holds Eq. (17) is fulfilled and yields after forming linear combinations of  $A_2$ ,  $B_2$ , and  $E$  symmetry,

$$TF^-(\gamma) = -C_2 F^-(\gamma). \quad (31)$$

Clearly, Eqs. (30) and (31) contradict each other. Thus  $T$  symmetry is always broken in all three cases in a robust way, i.e., independent of specific Hamiltonians and values for microscopic coupling parameters.

For completeness let us consider the case of an usual CDW without internal bond degrees of freedom. Eq. (9) reads for  $\mathbf{e}_j = 0$   $F^*(\mathbf{q}_l) = C_2 F(\mathbf{q}_l) = F(-\mathbf{q}_l)$ . Thus Eq. (18) is always fulfilled. Forming suitable linear combinations to get basis functions for irreducible representations we see that  $T$  symmetry is always unbroken in an usual CDW.

The above analysis showed that many of the symmetry allowed OPs break  $T$  symmetry. The finite imaginary part of these OPs generate circulating currents and space-dependent magnetic fields<sup>15,26</sup> which so far could not be observed<sup>27-29</sup>. Concentrating therefore on  $T$  conserving OPs there are none without fine-tuning in the case of a star with wave vectors along the diagonals. For the experimentally observed star with wave vectors along the crystalline axes there are three OPs which preserve  $T$  symmetry in a robust way. They have the symmetries  $A_1$ ,  $B_1$  and  $E$  and originate from the  $F^+$  manifold. In particular, the  $F^+(B_1)$  state with  $B_1$  symmetry seems to be a good candidate for the OP in underdoped cuprates<sup>8</sup>. Its bond charge pattern  $\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle$  is proportional to  $-\cos(qr_{iy})$  for  $\mathbf{e}_j = \mathbf{e}_1$  and  $\cos(qr_{ix})$  for  $\mathbf{e}_j = \mathbf{e}_2$  and is illustrated in Fig. 1. The color on each bond indicates the value for the corresponding bond charge. The pattern represents a simple bidirectional BOW state where the charges on the horizontal and vertical bonds vary only in one direction. Different ground states for underdoped cuprates have also been proposed, for instance, uniaxial BOW states with<sup>22</sup> or without<sup>8,22</sup>  $T$  breaking. Interesting is that the relevant OPs of the hot spot model of Ref.<sup>22</sup> are closely related to our manifolds  $F^+$  and  $F^-$  concerning point group and time reversal symmetries.

The OP deduced from experimental data in Ref.<sup>8</sup> uses a form for the OP which is based on the approximation

$$F(\mathbf{q}_l, \mathbf{e}_j) = e^{-i\mathbf{e}_j \mathbf{q}_l / 2} \sum_{\mathbf{k}} \Delta(\mathbf{k}) e^{i\mathbf{e}_j \cdot \mathbf{k}}. \quad (32)$$

This approximation is obtained from Eq. (3) by shifting the sum over  $\mathbf{k}$  by  $-\mathbf{q}_l/2$  and using for the matrix element the  $\mathbf{q}_l$  independent function  $\Delta(\mathbf{k})$ . From Eq. (9) follows that  $\Delta(\mathbf{k})$  has to be real. Inserting Eq. (3) into Eq. (2) and using Eq. (32) yields

$$\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \sum_l e^{-i\mathbf{q}_l(\mathbf{r}_i + \mathbf{r}_j)/2} \sum_{\mathbf{k}} e^{i\mathbf{e}_j \cdot \mathbf{k}} \Delta(\mathbf{k}). \quad (33)$$

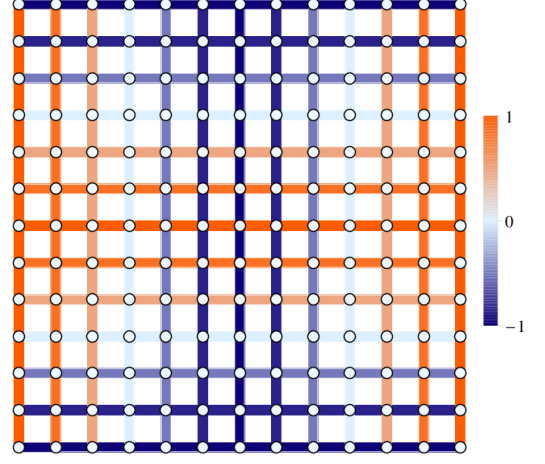


FIG. 1: (Color online) Bond charge pattern of the  $F^+(B_1)$  state using  $q = \pi/6$ .

The sum over  $\mathbf{k}$  is only nonzero if  $\Delta(\mathbf{k})$  is a linear combination of  $\cos k_x + \cos k_y$ ,  $\cos k_x - \cos k_y$ ,  $\sin k_x$  and  $\sin k_y$ . Inserting these functions into Eq. (33) and using the transformation rule of Eq. (4) shows that these patterns have  $A_1$ ,  $B_1$ , and  $E$  symmetries. For instance, for the  $B_1$  symmetry we have

$$\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \sum_l e^{-i\mathbf{q}_l(\mathbf{r}_i + \mathbf{r}_j)/2} \sum_{\mathbf{k}} e^{i\mathbf{e}_j \cdot \mathbf{k}} \Delta_0(\cos k_x - \cos k_y), \quad (34)$$

where  $\Delta_0$  is a real constant. The bond charge patterns of Eq. (33) are invariant if  $\mathbf{q}_l$  is transformed as  $\mathbf{R}^{-1}\mathbf{q}_l$  and  $\mathbf{e}_j$  is kept fixed. This transformation describes a permutation of the wave vectors of the BOW and does not correspond to an element of the point group  $C_{4v}$ . Invariance under this transformation represents an additional symmetry which we will call  $Q$  symmetry in the following. Applying  $T$  to Eq. (33) yields

$$T\langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle = \pm \langle c_{i,\alpha}^\dagger c_{j,\alpha} \rangle, \quad (35)$$

where the upper and lower sign holds for the  $A_1, B_1$  and  $E$  representations, respectively. The corresponding basis functions are therefore real or imaginary. Eq. (33) is identical with Eq. (S9) in Ref.<sup>8</sup>. This equation was used to analyse inelastic X-ray data in underdoped cuprates and it was concluded that, disregarding uni-directional modulations, the ground state has  $B_1$  symmetry<sup>8</sup>. The corresponding bond charges are proportional to  $-\cos(qr_{ix} + 1/2) - \cos(qr_{iy})$  for  $\mathbf{e}_j = \mathbf{e}_1$  and to  $\cos(qr_{ix}) + \cos(qr_{iy} + 1/2)$  for  $\mathbf{e}_j = \mathbf{e}_2$ , and yield a quite different pattern from that shown in Fig. 1.

As shown above the approximation Eq. (32) yields for each of the symmetries  $A_1$ ,  $B_1$ , and  $E$  just one OP. The charge patterns with  $A_2$  and  $B_2$  symmetries, discussed in section IV, no longer exist in this approximation. Eq. (32) also implies severe restrictions in the space of OPs. For instance, if  $\mathbf{q}_l$  and  $\mathbf{e}_j$  are perpendicular to each other

the functions  $F(\mathbf{q}_l, \mathbf{e}_j)$  become identical for  $l \neq j$ . Moreover, using the approximation Eq. (32), Eqs. (24) and (27) hold which means  $T$  symmetry for the  $A_1$  and  $B_1$  states and at the same time fine-tuning of OPs. It seems therefore preferable not to specialize  $F(\mathbf{q}_l, \mathbf{e}_j)$  as in Eq. (32) but to stick to the general form of the OPs and to use our previous general symmetry classification. We will restrict the discussion in the following to  $B_1$  states, but similar arguments also apply to the symmetries  $A_1$  and  $E$ .

In section IV we found that there are 3 different representations with  $B_1$  symmetry. The ground state OP  $F_0(B_1)$  will therefore be in general a linear combination of their basis functions, i.e.,

$$F_0(B_1) = \alpha F(B_1) + \beta \tilde{F}(B_1) + \delta F^+(B_1), \quad (36)$$

where  $\alpha, \beta$  and  $\delta$  are real numbers. The charge patterns form a two-fold manifold which is quite different from the case where Eq. (32) holds and only one OP exists. This difference is due to the constraints in the space of OPs introduced by the approximation Eq. (32). Next we simplify  $F_0(B_1)$  by requesting that it exhibits  $Q$  symmetry.  $F_0(B_1)$  then specializes unambiguously to  $\hat{F}(B_1)$  given by

$$\hat{F}(B_1) = \sum_{\mathbf{k}, l, j} \langle c_{\mathbf{k}+\mathbf{q}_l, \alpha}^\dagger c_{\mathbf{k}, \alpha} \rangle e^{i\mathbf{k}\mathbf{e}_j} (-1)^{j+1}. \quad (37)$$

Regrouping the terms to form irreducible basis functions we get

$$\hat{F}(B_1) = F(B_1) + \tilde{F}(B_1) - F^+(B_1). \quad (38)$$

Although  $\hat{F}(B_1)$  and Eq. (34) possess both  $Q$  and  $B_1$  symmetry and are unambiguously determined by these symmetries they are different. This can be seen from their behavior under time reversal. Eq. (34) is  $T$  symmetric according to Eq. (35). Applying  $T$  to Eq. (38) and using Eqs. (19) - (22) gives

$$T\hat{F}(B_1) = e^{-iq}\tilde{F}(B_1) + e^{iq}F(B_1) - F^+(B_1). \quad (39)$$

Thus  $\hat{F}(B_1)$  breaks in general  $T$  symmetry because Eq. (24) or the equivalent Eq. (28) are in general not fulfilled so that the first two terms on the right-hand side of Eq. (39) are not equal to  $\tilde{F}(B_1) + F(B_1)$ . According to our previous discussion equations like  $ReF(B_1) = Re\tilde{F}(B_1)$  are satisfied only for fine-tuned OPs which neglect the contribution from circulating currents associated with  $T$  breaking of  $F(B_1), \tilde{F}(B_1)$ , and  $\hat{F}(B_1)$ . In contrast to that  $F^+(B_1)$  is  $T$  symmetric without any restrictions. Besides of the most general OP of Eq. (36) there are three distinguished and simple possibilities for the ground state OP with  $B_1$  symmetry:

- (a) One is  $F^+(B_1)$  obeying  $T$  but not  $Q$  symmetry;
- (b) Another is given by Eqs. (37) and (38) exhibiting  $Q$  but breaking in general  $T$  symmetry;
- (c) State (b) with  $T$  symmetry due to fine-tuning; this

state is equivalent to Eq. (34).

Note that  $Q$  symmetry is in our case not an exact symmetry because the point group transformations change both the momenta of the BOW and the bonds at the same time. Thus (b) and (c) represent approximate states. In contrast to that state (a) is not  $Q$  symmetric but necessarily invariant under time reversal because this is a result of symmetry. Moreover, it is the only state which has this property. Since experiments seem to rule out circulating currents in underdoped cuprates<sup>27-29</sup> the ground state should be state (a) if fine-tuned states (i.e., states with restrictions not enforced by symmetry) can be ruled out.

## V. CONCLUSIONS

In conclusion, we have identified symmetry allowed bond OPs for any model with nearest neighbor interactions such as the  $t$ - $J$  model and studied their properties, in particular, with respect to time reversal. The obtained results are relevant for recently observed charge-ordered states in underdoped cuprates and their symmetries. The proposed OPs are more general than the variational Ansätze used in the past both in theoretical and experimental studies. Being based on rigorous group theoretical considerations our results are useful to design improved variational forms for the OP in microscopic calculations or to interpret experimental data.

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### Appendix A: Reducible and irreducible representations of the ground state.

It is well known that ground state wave functions form, disregarding accidental degeneracies, basis functions for irreducible representation of the symmetry transformations commuting with the Hamiltonian.<sup>25</sup> In this appendix we will study the question whether a similar statement is true for the ground state of a system described by a free energy functional and OPs.

Let us denote by  $F_0$  one of the OPs describing the ground state, i.e., which correspond to the minimum of the free energy.  $F_0$  can be represented as a linear combination of the  $F(\mathbf{q}_l, \mathbf{e}_j)$  and thus transforms in a well-defined way under point group transformations. Considering  $C_{4v}$  and applying its  $n=8$  transformations  $P_R$  to  $F_0$  we denote by  $R_i, i = 1, \dots, d$  the transformations which lead to linearly independent functions  $f_i = P_{R_i} F_0, i = 1, \dots, d$ . The functions  $P_{R_i} F_0, i = d+1, \dots, n$  can be written as linear combinations of the functions  $f_i$ . Denoting

the  $n$  point group operators by  $P_{R_\alpha}, \alpha = 1, \dots, n$ , we can write

$$P_{R_\alpha} F_0 = \sum_{i=1}^d f_i L_{i,\alpha}, \quad (\text{A1})$$

where  $L_{i,\alpha}$  is a matrix with  $d$  rows and  $n$  columns. Applying  $P_{R_\alpha}$  to  $f_i$  gives

$$P_{R_\alpha} f_i = P_{R_\alpha} P_{R_i} F_0 = P_{R_\alpha R_i} F_0 = \sum_{j=1}^d f_j L_{j,\bar{i}}. \quad (\text{A2})$$

$\bar{i}$  denotes the column of  $L$  which corresponds to the point group transformation  $R_\alpha \cdot R_i$ . For a fixed  $R_\alpha$   $\bar{i}$  runs over  $d$  columns of  $L$  which can be used to form a square matrix  $\Gamma(R_\alpha)$  with  $d$  rows and  $d$  columns. Eq. (A2) can now be written as

$$P_{R_\alpha} f_i = \sum_{j=1}^d f_j \Gamma(R_\alpha)_{j,i}. \quad (\text{A3})$$

Considering a product of two transformations  $R_\alpha$  and  $R_\beta$  we have

$$\begin{aligned} P_{R_\alpha R_\beta} f_i &= P_{R_\alpha} P_{R_\beta} f_i = P_{R_\alpha} \sum_{j=1}^d f_j \Gamma(R_\beta)_{j,i} = \\ &= \sum_{j,k} f_k \Gamma(R_\alpha)_{k,j} \Gamma(R_\beta)_{j,i} = \sum_k f_k (\Gamma(R_\alpha) \cdot \Gamma(R_\beta))_{k,i} \end{aligned} \quad (\text{A4})$$

On the other hand is

$$P_{R_\alpha R_\beta} f_i = \sum_j f_j \Gamma(R_\alpha R_\beta)_{j,i}, \quad (\text{A5})$$

so that

$$\Gamma(R_\alpha R_\beta) = \Gamma(R_\alpha) \cdot \Gamma(R_\beta). \quad (\text{A6})$$

Eqs. (A3) and (A6) establish that the matrices  $\Gamma(R_\alpha)$  together with the basis functions  $f_i$  form a representation of  $C_{4v}$ .

Assuming that no accidental degeneracy of the ground state is present the set of functions  $f_i$  form a basis for the degenerate OPs of the ground state. If  $d = 1$  there is only one linearly independent OP describing the ground state. Moreover, this OP must form a basis function for a one-dimensional irreducible representation of  $C_{4v}$ . Thus if the ground state is non-degenerate its OP must be one of the basis functions belonging to irreducible one-dimensional representations discussed in sections III and IV.

In the case  $d = 2$  there are two linearly independent OPs describing the degenerate ground state. They form a two-dimensional representation of  $C_{4v}$ . The two basis functions have the same free energy because they transform into each other by symmetry operations of the point group. Determining the trace of the associated representation matrices  $\Gamma_{i,j}$  there are two cases possible. The

representation is irreducible and the two basis functions transform according to the  $E$  representation. Or, the representation is reducible and a superposition of two different one-dimensional representations. In this case the free energy of the two-dimensional reducible representation may be lower or higher than that of the irreducible representations. This means that the case is not excluded that a two-fold degenerate ground state transforms according to a reducible and not an irreducible representation.

In order to illustrate the above statements we consider a simple free energy model without bond degrees of freedom and two real OPs,

$$\phi_1 = F(\mathbf{q}_1 + F(-\mathbf{q}_1)), \quad (\text{A7})$$

$$\phi_2 = F(\mathbf{q}_2 + F(-\mathbf{q}_2)). \quad (\text{A8})$$

$\mathbf{q}_1$  and  $\mathbf{q}_2$  are wave vectors of equal length along the  $x$  and  $y$  axis, respectively. Forming the combinations

$$\phi_A = (\phi_1 + \phi_2)/\sqrt{2}, \quad \phi_B = (\phi_1 - \phi_2)/\sqrt{2}, \quad (\text{A9})$$

yields basis functions  $\phi_A$  and  $\phi_B$  for a  $A_1$  and a  $B_1$  representation, respectively. We consider the following free energy functional,

$$F = \frac{a}{2}(\phi_A^2 + \phi_B^2) + \frac{1}{4}\phi_A^4 + \frac{g_B}{4}\phi_B^4 + \frac{g}{2}\phi_A^2\phi_B^2, \quad (\text{A10})$$

in the parameter range  $0 < g_B < 1$  and  $g \geq -\sqrt{g_B}$ . The underlying symmetry group is  $C_{4v}$ . The coefficient  $a$  is proportional to  $T - T_c$  and becomes negative below the transition temperature  $T_c$  to the BOW state. Note that the prefactor  $a$  is the same for both order parameters though the latter have different symmetries. In the usual terminology this corresponds to an accidental degeneracy. In our case this degeneracy is caused by the fact that the diverging susceptibilities in the normal state at  $\pm\mathbf{q}_1$  and  $\pm\mathbf{q}_2$  are related by point group operations. There is only one  $T_c$  for both symmetries  $A_1$  and  $B_1$ . Below  $T_c$  the OPs  $\phi_A$  and  $\phi_B$  will become finite and in general be also different due to the anharmonic terms in  $F$ .

Solving the extremal equations for  $F$  yields the following solutions:

$$(a) \quad \phi_A = 0, \phi_B = \pm\sqrt{\frac{-a}{g_B}}, \quad (\text{A11})$$

$$(b) \quad \phi_B = 0, \phi_A = \pm\sqrt{-a}, \quad (\text{A12})$$

$$(c) \quad \phi_A = \pm\sqrt{\frac{(-a)(g_B - g)}{g_B - g^2}}, \phi_B = \pm\sqrt{\frac{(-a)(1 - g)}{g_B - g^2}}. \quad (\text{A13})$$

The free energy Eq. (A10) is invariant under  $\phi_A \rightarrow -\phi_A$  and  $\phi_B \rightarrow -\phi_B$  which leads to additional degeneracies described by  $\pm$  in Eqs. (A11) - (A13). It is convenient to

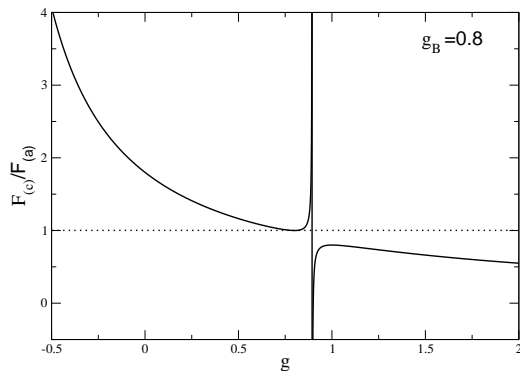


FIG. 2: Ratio of the free energies  $F_{(c)}$  and  $F_{(a)}$  as a function of  $g$  using  $g_B = 0.8$ . The dashed line corresponds to a ratio of 1.

take this degeneracy tacitly into account and to consider only half of the above solutions. Calculating also the corresponding free energies we find that (a) represents a basis function  $\phi_B$  of length  $\sqrt{\frac{-a}{g_B}}$  for a  $B_1$  representation with energy  $-\frac{a^2}{4g_B}$ . Similarly, (b) represents a basis function of length  $\sqrt{-a}$  for a  $A_1$  representation with energy  $-\frac{a^2}{4}$ . Finally, (c) consists of two degenerate basis functions with the components  $(\phi_A, \phi_B)$  and  $(\phi_A, -\phi_B)$ , respectively, where  $\phi_A$  and  $\phi_B$  are given by the expressions of Eq. (A13), omitting  $\pm$  in these expressions. The two basis functions yield a reducible two-dimensional rep-

resentation containing both  $A_1$  and  $B_1$  symmetries. The corresponding free energy is  $-\frac{a^2(1+g_B-2g)}{4(g_B-g^2)}$ .

Because of the assumption  $0 < g_B < 1$  the free energy of (a) is always lower than that of (b) so that the ground state is given either by (a) or by (c). Fig. 2 shows the ratio of the free energies of (c) and (a) as a function of  $g$  for  $g_B = 0.8$ . For  $g > \sqrt{g_B}$  the curve in Fig. 2 is always below 1. Thus the solution (a) has in this region the lowest free energy and describes the stable state. It is non-degenerate and its basis function belongs to an irreducible representation of  $B_1$  symmetry. For  $|g| < \sqrt{g_B}$  the curve in Fig. 2 is larger or equal to one. As a result solution (c) has the lowest free energy in this interval and describes a degenerate ground state. It is given by a two-dimensional reducible representation which consists both of  $A_1$  and  $B_1$  components. Our calculation shows that this two-dimensional reducible representation may have a lower free energy than the  $A_1$  and  $B_1$  components. Our calculation also demonstrates that the same basis functions may describe a degenerate or a non-degenerate ground state depending on the values of the coupling constants. This means that the degeneracy of a ground state described by a reducible two-dimensional representation is not enforced by symmetry as in the case of irreducible representations but depends in general on the values of the coupling constants. It thus can be considered to be accidental.

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